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PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Serial No.: 10/542,753
In re Application of:
Nobuhiro Yabunouchi et al.
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Confirmation No. 1609
Group Art Unit: 1713
Examiner: CHOI, LING SIU

For: Catalysts For Polymerizing Olefins And Process For Producing Olefin Polymer

DECLARATION UNDER 37 CFR 1.132

I, Shojiro TANASE, declare that:

1. I received a master degree from Nagaoka University of Technology in March, 1992. I have been employed since April, 1992 by IDEMITSU KOSAN CO., LTD. of 6-1, Yokoami 1-chome, Sumida-ku, Tokyo 130-0015 Japan, and had been engaged in research on catalysts for the polymerization of propylene, in the Polymer Research Laboratory of that company from April, 1992 to March, 2004 and Chemical Research Laboratory of that company from April, 2004 to September 2007. I have personal knowledge of the facts stated herein, except for these facts which are based on information and belief.

2. All statements made herein based on my own knowledge are true and all statements made on information and belief are believed to be true.

3. Experiment was conducted to establish the patentability of the claimed invention. The object of the experiment is to show that it is necessary to use an alkoxy-containing magnesium compound obtained by reacting metal magnesium, an alcohol and a halogen and/or a halogen-containing compound containing at least 0.0001 gram atom of halogen atoms per mol of the metal magnesium in order to obtain desired advantages of the present invention.

4. Experiment

A diethoxy magnesium compound was prepared in the same way as in Example 1 (1) of this U.S. Patent Application Serial No. 10/542,753 except that iodine was not used. Thereafter using the diethoxy magnesium compound, a solid catalyst component was prepared and propylene was polymerized in the same way as in Example 2 (1) and (2) of this U.S. Patent Application Serial No. 10/542,753.

Specifically the experiment was conducted as follows:

(1) Preparation of alkoxy-containing magnesium compound

A glass reactor having an internal volume of 6 liter and having a stirrer was fully flushed with nitrogen, and about 2,430 g of ethanol, and 160 g (6.6 mol) of metal magnesium were poured therein and allowed to react under heating and reflux with stirring until no hydrogen was generated from the system, to give a diethoxy

magnesium compound (average particle diameter; 540 μm). The diethoxy magnesium compound was grinded by a ball mill for 24 hours to give a diethoxy magnesium compound (average particle diameter; 68 μm).

(2) Preparation of solid catalyst component

A three-necked flask having an internal volume of 0.5 liter and having a stirrer was flushed with nitrogen, and 80 ml of dehydrated octane and 16 g of diethoxymagnesium prepared in the above (1) were poured therein. The mixture was heated to 40°C and 2.4 ml of silicon tetrachloride was added. After stirring for 20 minutes, 1.8 ml of diethyl n-butyl-malonate was further added. The solution was heated to 65°C and 77 ml of titanium tetrachloride was then dropped. The mixture was stirred for 2 hours at an internal temperature of 125°C, thereby carrying out a contacting procedure. Thereafter, it was fully washed with dehydrated octane. Titanium tetrachloride was added in an amount of 122 ml and stirred for 2 hours at an internal temperature of 125°C, thereby carrying out a contacting procedure again. Thereafter, it was fully washed with dehydrated octane to give a solid catalyst component.

(3) Propylene polymerization

An autoclave made of stainless steel having an internal volume of 1 liter and having a stirrer was fully dried and flushed with nitrogen, and 400 ml of dehydrated heptane was placed therein at room temperature. Further, 2.0 mmol of triethylaluminum, 0.25 mmol of cyclohexylmethyldimethoxysilane, and 0.0025 mmol in terms of Ti atom of the solid catalyst component prepared in the above (2) were added. Hydrogen was introduced up to 0.1 MPaG. Sequentially the

temperature and total pressure were raised to 80°C and 0.8 MPaG, respectively while introducing propylene. Polymerization was carried out for 1 hour.

Then, the temperature and pressure were decreased, and a reaction product was taken out and poured into 2 liter of methanol, thereby deactivating the catalyst. The product was removed by filtration and vacuum-dried to give polypropylene.

(4) Evaluation

The catalyst and polypropylene obtained were evaluated in the same way as in Example 1 as follows:

(1) Catalyst activity: A solid catalyst component produced was fully dried, exactly weighed and fully decomposed with a 3N sulfuric acid. Insoluble materials were removed by filtration. To the filtrate were added phosphoric acid as a masking agent and 3% hydrogen peroxide solution, thereby developing color. The absorption of the colored solution at 420 nm was measured by FT-IR to determine a Ti concentration. The amount of supported Ti in the solid catalyst component was calculated using the Ti concentration and the catalyst activity per gram of titanium was calculated on the basis of the amount.

(2) [η] : Measured in a tetralin solvent with a VMR-053 model automatic viscometer available from Rigo Co., LTD.

(3) [mmmm] : An olefin polymer obtained was dissolved in a 90:10 (volume ratio) mixture solvent of 1,2,4-trichlorobenzene and

deuterobenzene, and isotacticity was determined on the basis of signals of methyl measured at 130°C by a proton complete decoupling method using ^{13}C-NMR (trade name: LA-500, available from JEOL).

[mmmm] is an isotactic fraction in pentad units of a polypropylene molecule chain determined on the basis of ^{13}C-NMR spectrum as proposed by A. Zambelli et al on page 925 of Macromolecules, Vol. 6 (1973).

Further, the method of assignment of peaks of ^{13}C-NMR spectrum was according to the assignment proposed by A. Zambelli et al. on page 687 of Macromolecules, Vol. 8 (1975).

(4) Average particle diameter of alkoxy-containing magnesium compound: A magnesium compound was suspended in a hydrocarbon, and the magnesium compound in this state was measured for particle diameters by a light transmission method. Particle diameter distribution determined by the measurement was plotted on a logarithmic normal probability paper, and a 50 % particle diameter was taken as an average particle diameter (D_{50}).

(5) Particle state of polyolefin powder (average particle diameter, rough powder amount and fine powder amount: Particle diameter distribution measured with a sieve was plotted on a logarithmic normal probability paper, and a 50 % particle diameter was taken as an average particle diameter. The amount of rough powder whose diameter is 2,830 μm or more, and the amount of fine powder whose diameter is 250 μm or less were obtained.

Table shows the results of the above experiment and the data

of Example 2 for reference. As stated above, the experiment is different from Example 2 only in preparation of a diethoxy magnesium compound.

Table

	Catalyst activity (kg-pp/g-Ti)	Polymer properties		Morphology			
		[η] (dl/g)	[mmmm] (%)	Mg Compound Average particle diameter (μm)	Polymer Average particle diameter (μm)	Polymer Rough powder amount (%)	Polymer Fine powder amount (%)
Experiment	165	0.94	92.6	68	670	15.8	2.0
Example 2	252	0.98	93.4	62	1100	0.3	3.6

The above table shows that a difference in preparation of a diethoxy magnesium compound causes a great change in catalyst activity and properties of polymer. The present invention uses an alkoxy-containing magnesium compound obtained by the specified method. Use of the alkoxy-containing magnesium compound enables the advantages of the present invention.

I declare under penalty of perjury under the laws of the United States that the foregoing is true and correct. I acknowledge that willful false statements and the like are punishable by fine or

imprisonment, or both and may jeopardize the validity of the application or any patent issuing thereon.

Date: Oct. 26, 2007

Shojiro Tanase

SHOJIRO TANASE